Journal of Cultural Heritage 16 (2015) 401–412

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Original article

Influence of manufacturing parameters on the crackling process of ancient Chinese glazed ceramics☆

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A R T I C L E I N F O

Article history:
Received 13 April 2014
Accepted 3 October 2014
Available online 1 December 2014

Keywords:
Crack
Glaze
Ceramic
Chinese wares
Song Dynasty

A B S T R A C T

Guan and Ge wares, produced during the Song Dynasty (960–1279 AD), hold a very special position in Chinese ceramic history because of their aesthetical qualities with a prominent crackle as their only decorative feature. The aim of this inter-disciplinary research is to understand the formation of crack patterns in ancient Chinese glazed ceramics in order to gain knowledge on the manufacturing process. We propose a new approach based on a time-scale investigation of the crackling process and of the cracks morphology obtained on glazed ceramic model systems synthesized under controlled conditions. In order to establish a link between macroscopic and microscopic properties, EDXRF, XRD and SEM-EDX analyses have been performed. Our results show that the relative glaze-body thickness and the firing temperature and atmosphere are key factors to control the crack patterns morphology.

1. Introduction

Glaze crackling is a frequently occurring phenomenon in ancient Chinese wares, however, it is only during the Song Dynasty (960–1279 AD) that people especially the Nobles and the Royal Family began to pay more attention to glaze crackling and valued it at an aesthetic level [1]. Guan and Ge wares, produced at this period, usually have prominent crackles as their only decorative feature. Because of their popularity, they were extensively copied under the following Yuan (1279–1368 AD), Ming (1368–1644 AD) and Qing (1644–1911 AD) dynasties [2].

Guan ware often has relatively large cracks whereas Ge ware is usually characterized by a double crackle network with lines varying in width and color: the major crackle is deliberately stained black and the minor crackle appears gold-brown [2]. The shape, size, colour and decoration of Guan and Ge wares have been well studied and classified according to the period and site of production [3,4]. The global chemical composition of their glazes and bodies is also well known [5–8]. However, many questions remain on the origin and on the manufacturing techniques of these wares.

Whereas cracks in varnishes and paint have been the interest of many researchers in the field of Cultural Heritage these last decades, cracks in glazed ceramics have been almost totally neglected [9,10]. However, in vitreous materials, the morphology of crack patterns is a direct marker of the object history since it depends on the glaze-body properties, which comprises the chemical composition, the microstructure, the mechanical properties and the geometry (thickness, curvature, stratigraphy, etc.). This paper aims to demonstrate that new information on ancient Chinese crackled wares can be provided through the comparative study archaeological samples and model systems with controlled geometry and physico-chemical properties.

Despite the apparent complexity of crack patterns, the formation of cracks is controlled by some rules. Cracks are produced in response to a mechanical frustration that occurs during the cooling, when the glaze solidifies and binds to the ceramic body with different thermal expansion [11]. Our approach focuses on the effect of the manufacturing parameters on the crack formation process and on its resulting morphologies of crack patterns.

2. Material and methods

2.1. Archaeological samples

2.1.1. Material

Twenty-three samples of crackled glazed ceramics dating from the Song and Yuan Dynasties have been studied. These shards

☆ “Best presentation of a young scientist at the 12th European Meeting on Ancient Ceramics (EMAC2013)”.

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http://dx.doi.org/10.1016/j.culher.2014.10.003
1296-2074/

Table 1
Weight of the raw materials used to synthesized 100 g of Song-like and Yuan-like glazes.

Raw materials	Feldspar	Kaolin	Calcite	Quartz	Iron oxide
Song-like glaze	29.9	18.9	20.0	30.4	0.8
Yuan-like glaze	59.5	7.7	13.2	18.7	0.9

belong to the collection of the Key Scientific Research Base of Ancient Ceramics, Shanghai Institute of Ceramics. They were excavated from archaeological sites of Jiaotanxia and Laohudong, and collected from Fenghuang Hill. We have selected these samples according to the variety of crackle patterns. They have different shapes and sizes, as well as body and glaze colorations and thicknesses [12]. It implies that various raw materials, manufacturing and firing conditions have been used. The inner and the outer glazes can also differ notably within one sample, so they are considered as independent glazes, ranging the total number of studied glazes to forty-six.

2.1.2. Imaging process

Contrary to glazed ceramic model systems, the morphology of cracks patterns of archaeological samples is not always clearly visible at naked eyes. Indeed, most of the glazes are constituted by very thin translucent cracks and the glaze surface is often weathered or dirty. In a previous paper, we have demonstrated that D-Stretch imaging process can be a powerful tool for researchers working on crackled ceramics since it enhances crackle network and brings out fine cracks that are invisible to naked eye due to the alteration of the surface (Fig. 1) [12].

We use the methodology described in [12] to enhance the crack patterns morphology. D-Stretch is used directly on digital camera images of archaeological samples [13]. Then, we can obtain statistical data on the number of domains, their size, shapes, distributions, etc. using ImageJ Analyze particle command [14]. An appropriate description of the crack pattern, based on the hierarchy and space-dividing properties, can be approach using the characteristic distance between cracks L as defined by Bohn et al., 2005 [15]: $L = \sqrt{(A/N)}$ where A is the observed area and N the number of domains in the pattern. We use this parameter to characterize crack morphology in all the samples studied.

2.2. Glazed ceramic model systems

2.2.1. Samples preparation

All model samples are made using one type of alumino-silica clay body coming from Zhejiang province. The clay is flattened to even thickness using slab roller equipment. Bodies of 2, 4 and 8 mm thickness before firing are prepared. Rectangular samples of equal sizes ($5 \times 6\,cm^2$) are sliced and kept for drying at room temperature for three days before being biscuit fired at low temperature (500–800 °C, 2 h). After biscuit firing, the different body thicknesses are respectively of 1.7, 3.8 and 6.5 mm. We have chosen to work on flat geometry samples in order to have a controlled thickness. Note that the curvature of the sample may very probably have an impact on the characteristics of the cracks since the repartition of the constraints will not be identical in every points of the system. Also depending on the curvature, the viscosity of the glaze, and the way the sample is placed in the furnace (vertically, horizontally, etc.), the glaze/body thicknesses can vary during the firing/cooling process.

Song-like and Yuan-like glazes with compositions close to archaeological samples are prepared using the appropriate amounts of raw materials: kaolinitic clay, feldspar, calcium carbonate and silica powder (Table 1). In order to apply the glaze on the body substrate, a fluid preparation is made by mixing the raw materials with an aqueous solution. All the raw materials – but the silica powder – are mixed together with 200 mL of water and milled for 9 min (ZrO_2 milling ball, KELI CERAMICS equipment). The adequate amount of silica powder is then added to the mixture and the whole is mixed together for 1 min at the same speed.

To apply the glaze on the body, it is believed that early Chinese technique was to tie a silk cloth over the end of a bamboo tube and then blow the liquid glaze from the tube through the cloth [5]. In order to conserve the idea of a blowing technique, glazes are applied using a spray gun linked to a vacuum pump. This technique allows obtaining homogeneous layers of glaze with a controlled and even thickness. For every layer of 0.2 mm, the applied glaze has been dried for 15 min in the oven in order to insure the good adhesion of the glaze to the body. Samples with final glaze thickness of 1, 1.5, 2, 3, 4 and 5 mm before firing have been prepared.

Samples are placed in the electric furnace and the temperature is raised slowly up to the firing temperature during 4 h. When the firing temperature is reached, samples are then fired at constant temperature during 30 min. Firing temperatures in the range of 1180 until 1250 °C are tested. In the same way, as potters did during the ancient dynasties, the samples are cooled slowly from firing temperature to room temperature before taking them out of the furnace (~24 h). In the same furnace, samples can be fired under reducing atmosphere by isolating them in a closed refractory box containing a reducing agent (coke). For all the experiments, the temperature is measured using ring temperature (Ferro rings #238).

2.2.2. Imaging process

In order to follow the dynamic of the crack formation and propagation, pictures of glazed ceramic model samples are taken at regular time intervals with fixed lighting and stable room temperature. Pictures are recorded using a Nikon DX 3200 camera and edited using GIMP 2.8 software [16]. For each picture, new cracks are traced and the geometry of the fractures is observed. The evolution of crack domains and their characteristics (number, size, distribution, etc.) can be followed in a time-scale (Fig. 2). The characteristic length of crack domains L defined in Section 2.1.2 is calculated and its evolution is followed at regular time intervals in all the model samples studied. We consider the crackling process has reached the kinetic equilibrium when the number of new cracks formed seems not to change with time anymore. In the first step of the project, we have measured the number of crack domains but have also recorded their respective geometry (area, perimeter, width, height, circularity etc.) [12]. However, due to the limited time, and to the apparently limited influence of these parameters on the cracks formation process – in comparison with the number of domains – we have chosen to focus on the study of the variation of the L parameter with the manufacturing process.

2.3. Chemical and structural analyses

Glaze and body compositions are analyzed by energy-dispersive X-ray fluorescence (EDXRF Eagle-III μ-Probe, USA, 40 kV, 15 min). The body analysis is performed on a polished cross-section surface, and the glaze analysis was performed on a natural external surface. Four measurements are carried out and the average is adopted for each analysis.

The samples are observed by optical microscope (KEYENCE instrument, VHX2000, Japan, magnification × 100–300) in order to measure the glaze and body thickness, observe cracks orientation and bubbles or crystals distribution.

Microstructure and micro-area composition is studied using scanning electron microscope (SEM; JEOL JXA 8100, Oxford Instruments, 20 kV). Backscattering imagery (BSE) is used and semi-quantitative analyses of the elemental compositions of vitreous